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A SWEEP GAS FACILITY FOR FISSION GAS RELEASE STUDIES AT THE NASA PLUM BROOK REACTOR

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16. Abstract

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SUMMARY

A sweep gas facility for measuring fission gas release rates from power producing nuclear fuel specimens has been constructed and used at the NASA Plum Brook Reactor Facility.

The helium sweep gas flow system and its supporting equipment are described. A sodium iodide scintillation detector system is used for online fission gas isotope analysis. Bottle samples of the sweep gas are easily obtained for more detailed laboratory analysis. The fuel specimen position in the reactor test hole can be manually or automatically adjusted to compensate for changes in the thermal neutron flux. The sweep gas facility has operated satisfactorily in tests with mockup and actual fuel specimens up to a current operating limit of 300 watts fission heat generation rate.

INTRODUCTION

The mechanism of fission gas release from nuclear fuels is not understood well enough to efficiently design fuel elements for space power reactors. The amount of fission gas release determines the extent of fuel swelling and gas pressure buildup in the fuel element. To obtain information about the fission gas release behavior of candidate fuels for space power reactors, a sweep gas facility was constructed at the NASA Plum Brook Reactor Facility.

This sweep gas facility provides a means for measuring and monitoring the activities of fission gases released from power producing fuel specimens. The release rates of these gases can then be determined as a function of time and fuel operating conditions.

This report describes the sweep gas facility, its capabilities, and some of its operating characteristics as determined during actual operation.

FACILITY DESCRIPTION

The sweep gas system is basically a helium flow system which provides a means of monitoring the fission gas release from power producing fuel samples. A simplified flow schematic of the sweep gas system is shown in figure 1. Bottle supplied helium is piped through a gas dehumidifier-purifier, flowmeter, and an irradiation capsule containing a fuel specimen. Gaseous fission products released from the irradiated fuel sample are swept out of the capsule by the helium through a copper wool filter (to remove iodine) and through a sodium iodide (NaI) scintillation detector source where the activity is measured. After flowing past the detector, the gases are stored in a large 11.3 cubic meter (400 ft^3) tank for intermediate holding and radioactive decay before release to the reactor off-gas system.

The general locations of the sweep gas system components at the Plum Brook Reactor (PBR) are shown in figures 2 and 3. The gas supply bottles and dehumidifier-purifier are located at the lower level of the reactor containment vessel dry annulus. The in-core portion of the facility (lead train, capsule, and positioning device) is located in one of the reflector test holes (RA-5) as shown in figure 2. The gas lines, as well as the receiver tank in quadrant A, are located under water for radiation shielding. The cabinet which contains a major portion of the facility support equipment is located on a catwalk in the dry annulus. The NaI detector station and flow system calibration panel are located adjacent to the equipment cabinet. The equipment cabinet contents, scintillation detector station, and calibration panel are shown in more detail in figure 3.

The control console and most of the electrical equipment associated with the facility are in the experiment control room annex, just outside the containment vessel.

The more important sweep gas facility components, design features, and operating capabilities are discussed in the following sections.

GAS FLOW SYSTEM

The gas flow system for this facility (refer to fig. 1) is designed to flow highly purified helium at a controlled flow rate past the fuel specimen and then out of the capsule to the counting and sampling equipment. Because of safety requirements, all major portions of the gas lines carrying fission gas are enclosed by a higher pressure static secondary gas system.

Various auxiliary operations, such as purging or evacuating selected sections of gas line, can be performed without disrupting the normal gas flow.

Helium supply system. - The helium supply for this system consists of commercially purified helium in high pressure bottles connected to an automatic pressure regulating manifold. Pressure switches in the manifold change bottle banks automatically when bottle pressure drops below a 2.2×10^{6} newton per square meter (315 lb/in.^{2}) set point. The manifold output pressure of 0.69×10^{6} newton per square meter (100 lb/in.^{2}) is maintained constant at all times.

As noted on the flow schematic (fig. 1), some of the supply bottles contain a mixture of helium plus 3 percent argon. This mixed gas can be used to monitor the neutron flux in the test capsule by means of the argon activity.

When necessary, gas purity is maintained through the use of an online purifier. This unit removes trace quantities of impurities from the bottled gas so that total impurity content is in the low ppm range. In addition, a molecular sieve further downstream ensures that essentially all the moisture has been removed from the helium (<1 ppm) before it enters the capsule.

Flow control system. - The gas flow rate can be varied from 0.10 to 1.0 cubic centimeter per second (0.0061 to 0.061 in.^{3}/sec) using an air-actuated flow control valve mounted in the equipment cabinet valve canister (fig. 3). The valve canister provides double containment for the valves and lines within the "double-dashed" lines in figure 1. The system flow volume from capsule to scintillation detector is approximately 1130 cubic centimeters (69.0 in.^{3}) so the inline travel time can be varied from 1130 to 11 300 seconds (approx. 19 to 190 min).

The flow control system also includes a flowmeter and supporting pressure and flow calibration equipment mounted on the calibration panel adjacent to the equipment cabinet (fig. 3).

It was necessary to shield certain components in and around the dry annulus equipment cabinet. Most of the in-cabinet equipment (principally the valve canister and adjacent sweep gas lines) is shielded with up to 10 centimeters (4 in.) of lead.

IN-TANK SYSTEM

The portion of the sweep gas system located inside the reactor tank consists of a lead train containing the capsule, iodine filter, and solenoid valve assembly (detailed in fig. 4) plus a capsule insertion device (fig. 6). The entire lead train is replaced as a unit with each new fuel specimen. The connection to the permanently installed portion of the sweep gas system is made at the top of the reactor tank by means of double end shutoff, quick-connect couplings.

Irradiation capsule. - The irradiation capsule presently being used is shown in figure 5. It is designed to irradiate small diameter fuel cylinders contained in a vented pin.

The fuel sample is contained in a cylindrical molybdenum-5 percent titanium-0.08 zirconium (TZM) pin. The upper end of the pin is vented to permit the escape of fission gases into the helium sweep gas. The pin is axially supported and centered in a helium heat conduction gap by means of the thermocouple sheathing tube (molybdenum) attached at each end of the pin.

Test temperatures of interest are maintained by the fission heat generated in the fuel pin. This heat is conducted radially outward through the pin wall, the gas conduction gap, the copper conduction ring, and finally through the capsule wall to be transferred by forced convection to the reactor primary cooling water. The fuel pin fission heat generation rate is currently limited to a calculated value of 300 watts. This value is presently considered to be the upper limit for safe operation.

The gas conduction gap width controls the fuel sample temperature for a given heat generation rate in any given fuel pin. It is varied from capsule to capsule depending on the pin temperature - power characteristics desired. The gap width is altered by changing the inside diameter of the copper ring.

Copper is used to transfer heat by conduction to the stainless steel capsule wall. This transfer method leads to greatly reduced radial temperature gradients and thermal stresses in the capsule. Since the thermal expansions of the copper and stainless steel match at their interface, differential expansion stresses are minimal. The copper is gold brazed to the stainless steel using an 82 percent gold - 18 percent nickel braze.

The sweep gas inlet tube is notched as shown in figure 5. Therefore, the major portion of the sweep gas flow is bypassed around the gas conduction gap. This condition assures the virtual elimination of forced convection heat transfer in the gap, and the capsule heat-transfer characteristics are then independent of gas flow rate.

The irradiation capsule contains three high temperature thermocouples: tungsten - 26 percent rhenium/tungsten - 5 percent rhenium, insulated with high purity BeO, and molybdenum sheathed. Two 1.59-millimeter- (0.062-in.) diameter sheath thermocouples force fit into holes at the ends of the fuel pin are used to suspend it in the gas gap. A 1.12-millimeter- (0.044-in.-) diameter sheath thermocouple is set in a slot machined in the TZM clad. This thermocouple senses the clad temperature midway along the pin.

The capsule flow shroud channels the reactor coolant past the outer surface of the capsule. It is required to maintain adequate coolant velocity at the capsule heat transfer surface adjacent to the fuel pin. (A flow shroud would not be required for a larger diameter capsule, since it would more closely approach the reactor test hole in size.)

Iodine filter. - Iodine is one of the fission-produced gases swept out of the capsule by the helium, but its presence in the sweep gas can lead to two problems. One is the health safety problem which would be encountered on accidental release of the iodine to the atmosphere. The second problem is that the iodine would tend to condense along the length of the sweep gas tube walls and thus result in a linear source of xenon. This would cause difficulties in calculating the xenon release rate based on downstream activity measurements.

To solve both of these problems, a pressed copper wool filter was installed just downstream of the capsule (fig. 4). Tests have indicated that a 5-centimeter (2 in.) long plug of pressed copper wool will remove over 99.5 percent of the iodine passing through as a gas. Hence, since almost all of the iodine carried out of the capsule is trapped and held in this filter, further travel is prevented.

Lead train. - The thermocouple leads and sweep gas lines are brought out of the capsule through a rigid support rod to the filter housing. From here the leads pass through a 1.9-centimeter- (0.75-in.-) diameter flexible hose and solenoid valve assembly to a junction box at the top of the reactor pressure vessel. The solenoid valve assembly affords a means of purging the gas lines and isolating the capsule prior to disconnecting the lead train after test. The rigid support rod provides the connecting link to the capsule positioning device described in the next section. The flexible hose which provides the secondary containment for the sweep gas lines, is required to compensate for movement of the capsule positioning device.

Capsule positioning device. - The capsule is positioned in the reactor test hole by means of a vertical adjustable facility tube (VAFT). This is a standard positioning device used at the Plum Brook Reactor.

A typical VAFT is shown in figure 6. The basic drive consists of a push-pull cable connected at the top through a work gear actuator to a reversible electric motor. The lower end of the push-pull cable is connected to a carriage rod, which in turn is connected to the capsule support rod.

Total VAFT movement is 38.1 centimeters (15.0 in.), and the maximum speed is 3.8 centimeters (1.5 in.) per minute.

VAFT withdrawal action can be either manual or automatic. Automatic withdrawal was deemed necessary because of the thermal neutron flux characteristics of the reactor test hole used. The thermal flux increases with time as the control rods are withdrawn during the course of the reactor cycle. The VAFT slowly withdraws to keep the fuel pin temperature constant.

VAFT withdrawal is initiated when a controlling fuel pin thermocouple exceeds a high temperature set point. A pulse timer - electric timer combination starts the VAFT motor for the few seconds needed to decrease the temperature to just below the set point.

There is no automatic VAFT insertion.

ACTIVITY MONITORING SYSTEMS

As shown in figure 1, the gas leaving the capsule flows past a geiger detector and on through a valve canister where it can be routed through a scintillation detector source and a sampling station.

Geiger Detector

The geiger counter is used to determine the gross fission gas activity in the sweep gas line between the capsule and the scintillation detector. The geiger tube located in water-filled quadrant A (fig. 2) is used to give advance warning of any abnormal release of fission gas.

Valve Canister

The canister houses and provides double containment for the various solenoid valves which permit flow through the scintillation detector source, the sampling station, or directly to storage (refer to fig. 1).

These solenoid valves are also used as blocking valves so that some sections of the sweep gas system can be purged with clean helium or evacuated using the compressor without disturbing the gas flow through the capsule. (All the canister valves are not shown in the simplified flow schematic (fig. 1).) Purging operations are required to empty the source probe prior to obtaining the scintillation detector system background count rate and before any gas lines are disconnected, such as for removal of sweep gas samples.

Scintillation Detector and Source

A scintillation detector system is used for online fission gas isotope analysis. The system consists of a source (a 1-cm^3 volume through which sweep gas can flow), a detector (a 7.6 by 7.6 cm NaI crystal mounted on a movable platform), and a multichannel analyzer.

The 1 cubic centimeter source (fig. 7) is part of the doubly contained sweep gas flow system, so that sweep gas when flowing through it is surrounded by 1.0×10^6 newton per square meter (150 lb/in.^{2}) helium. The source is axially alined with the scintillation detector as shown in figure 3.

The NaI detector is track mounted so that a reasonably high counting rate can be maintained even when the fission gas release rate at the fuel varies by orders of magnitude. At low release rates, the detector is moved toward the source; at high release rates, it is moved away.

Ambient temperatures at the detector location can climb to over 32^{o} C (90^{o} F). Therefore, to guard against changes in detector efficiency, a small refrigerator is used to provide some cooling of the crystal and photomultiplier.

Sample Station

The sample station assembly shown in figure 8 is used to take samples of the sweep gas for radioactive isotope analysis. The germanium detector used to analyze these samples has a much higher resolution than the online NaI scintillation detector. This higher resolution, coupled with the ability to more effectively eliminate background counts, enables us to obtain a more detailed gamma energy spectrum. As a result, counting precision is better, and more isotopes can be identified with the gas samples than with the online NaI detector system. The general location of the sample station is indicated in figure 3.

Three sizes of aluminum containers are used to take these samples. The general activity level of the sweep gas determines which size (either 1, 10, or 50 cm^3) bottle is used. The smallest container is for the higher activity samples. Gas activities can reach ~10 millicurie per cubic centimeter.

The gas sample containers are not doubly contained, so any leak would result in the release of radioactive gas. Therefore, special precautions must be taken during sampling. The sampling procedure includes leak testing the sample bottle immediately before use and also purging and evacuating the quick connect couplings before removing the sample.

GAS STORAGE AND CONTAINMENT SYSTEM

Storage Tank

All the gas which flows through the system is temporarily stored in an 11.3 cubic meter (400 ft^3) double walled tank to permit radioactive decay before discharge to the existing Plum Brook systems for handling radioactive gas.

Double Containment

Almost all of the primary gas lines which carry fission produced gases are enclosed in a secondary helium system maintained at a pressure of 1.0×10^6 newton per square meter (150 lb/in.^{2}), which is a factor of 1.5 higher than the nominal sweep gas pressure. This higher pressure secondary containment concept minimized possible radioactive gas release to the atmosphere.

The only parts of the primary system not doubly contained in the higher pressure secondary system are the sampling station (fig. 8) and the compressor. However, both of these components have special safeguards against fission gas leakage. The compressor contains a double O-ring sealed, double diaphragm with a pressure monitor on the space between the diaphragms. The sampling station is leak tested before every use.

INSTRUMENT CHANNELS

Main Data Channels

Most of the data readout and electrical control equipment for this facility is located in one of the Reactor Facility experiment control rooms. This equipment, mounted in four standard racks, includes (1) the multichannel analyzer system, (2) solenoid valve controls, (3) VAFT controls, and (4) gas flow controls.

Strip chart recorders are used to monitor the fuel pin end temperatures, gas flow rate, geiger detector count rate, and secondary containment pressure. All of these, as well as several sweep gas pressure measurements and the fuel pin side temperature, are available as digital readouts through the Plum Brook EDLAS (Experiment Data Logging and Alarm System) system.

Safety Channels

A basic design criteria of the experiment was to limit operating parameters so that in the event of credible accidents damage would be limited to the experiment itself (i.e., not cause damage to the reactor). However, to protect the experiment and personnel, automatic alarm action is provided on high fuel pin temperature, high or low system pressures and flow, and high fission gas activity (i.e., on geiger detector count rate).

The basic alarm actions are (1) to stop the production of fission products by withdrawing the VAFT and (2) to stop the sweep gas flow to minimize the movement of high activity fission gas.

IRRADIATION ENVIRONMENT

The unperturbed thermal neutron flux in the RA-5 reactor test hole varies with vertical position from approximately 3×10^{14} to approximately 3×10^{12} neutrons per square centimeter - second (fig. 9). (The thermal neutron flux measurements were made in the Plum Brook mockup reactor.) At the typical fuel pin locations (27.3 to 33.0 cm (10.8 to 13.0 in.) above the core center line) the flux is $\sim1.5\times10^{14}$ neutrons per square centimeter - second (unperturbed). During a reactor cycle the flux at a given position increases by about a factor of 2 due to movement of the reactor control rods.

Gamma heating rates vary between 6 and 1 watts per gram (measurements made using mockup reactor) at the typical capsule positions. The nominal fuel pin gamma heating rate is 3 watts per gram.

GENERAL FACILITY OPERATING EXPERIENCE

As of October 1970, the sweep gas facility had been used for three capsule tests. The first test used a complete but unfueled capsule - fuel pin assembly; the second and third tests were with capsules containing enriched UO_2 (0.13 g) and enriched UN (0.24 g), respectively. In all these tests the general operation of the facility was satisfactory.

The purpose of the run with the unfueled capsule was to determine some of the operating characteristics of the facility as well as to demonstrate facility reliability without fission product contamination to complicate any facility modification that might be required. The results of some of the system operating characteristics tests are now discussed.

(1) The sweep gas travel time from the capsule to the 1 cubic centimeter scintillation detector source was determined as a function of gas flow rate by timing the arrival of argon-41 (A-41) activity pulses. The activity pulses were produced in the gas (helium plus 3 percent argon) by changing the VAFT position and, hence, changing the neutron flux at the capsule. The activity pulses traveled downstream and were easily observed at the scintillation detector. An example of the data obtained is shown in figure 10. The flow volume calculated from these travel times is $\sim$1130 centimeters^3.

(2) Actual release rates of fission produced gases will be calculated from measured gas activities using

$$R = \frac{\dot{Q}Ae^{\lambda\tau}}{\lambda}$$

where

R release rate, atoms/sec

$\dot{Q}$ volumetric flow rate, cm^3/sec

A activity of source gas, disintegrations/sec-cm^3

λ decay constant, sec^{-1}

τ travel time, sec

This relation was verified by measuring the A-41 activity as a function of gas flow rate at constant flux. The results are shown in figure 11. The variation with flow rate is as anticipated.

(3) The activity of A-41 in the sweep gas was determined by the online scintillation detector and compared to the activity found by analysis of gas samples. The results showed a reproducible difference of ~3 percent between the two methods. (A 5-centimeter NaI scintillation detector was in use at this time.)

Facility operation during the fueled tests was satisfactory. Fission-produced gases were observed at the scintillation detector, and samples of these gases were easily taken for more detailed analysis. A representative fission gas gamma-ray energy spectrum obtained using the scintillation detector is shown in figure 12. Five fission gas isotopes can be identified in this spectrum. Using a Ge(Li) detector to analyze the gas samples gives much better resolution. A representative spectrum is shown in figure 13.

CONCLUDING REMARKS

A sweep gas facility has been built and operated at the NASA Plum Brook Reactor Facility. Operation has been satisfactory with mockup and actual fuel specimens up to a current operating limit of 300 watts fission heat generation rate.

The results of tests performed with an unfueled capsule demonstrated facility reliability during extended operation. These tests also employed an argon activation technique to measure system travel time as a function of gas flow rate and to verify an equation to be used to calculate fission gas release rate from measured activity and flow rate.

As seen in figure 10, pulse changes in the A-41 generation rate produced relatively well defined changes in the A-41 count rate at the scintillation detector. This indicates that mixing in the gaslines is minimal, so it should be possible to follow some of the changes in fission gas release rates after changes in fuel pin position, flux, and power.

The tests on two fueled specimens showed that the facility can be used to measure steady-state fission gas release rates of several krypton and xenon isotopes.

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National Aeronautics and Space Administration,
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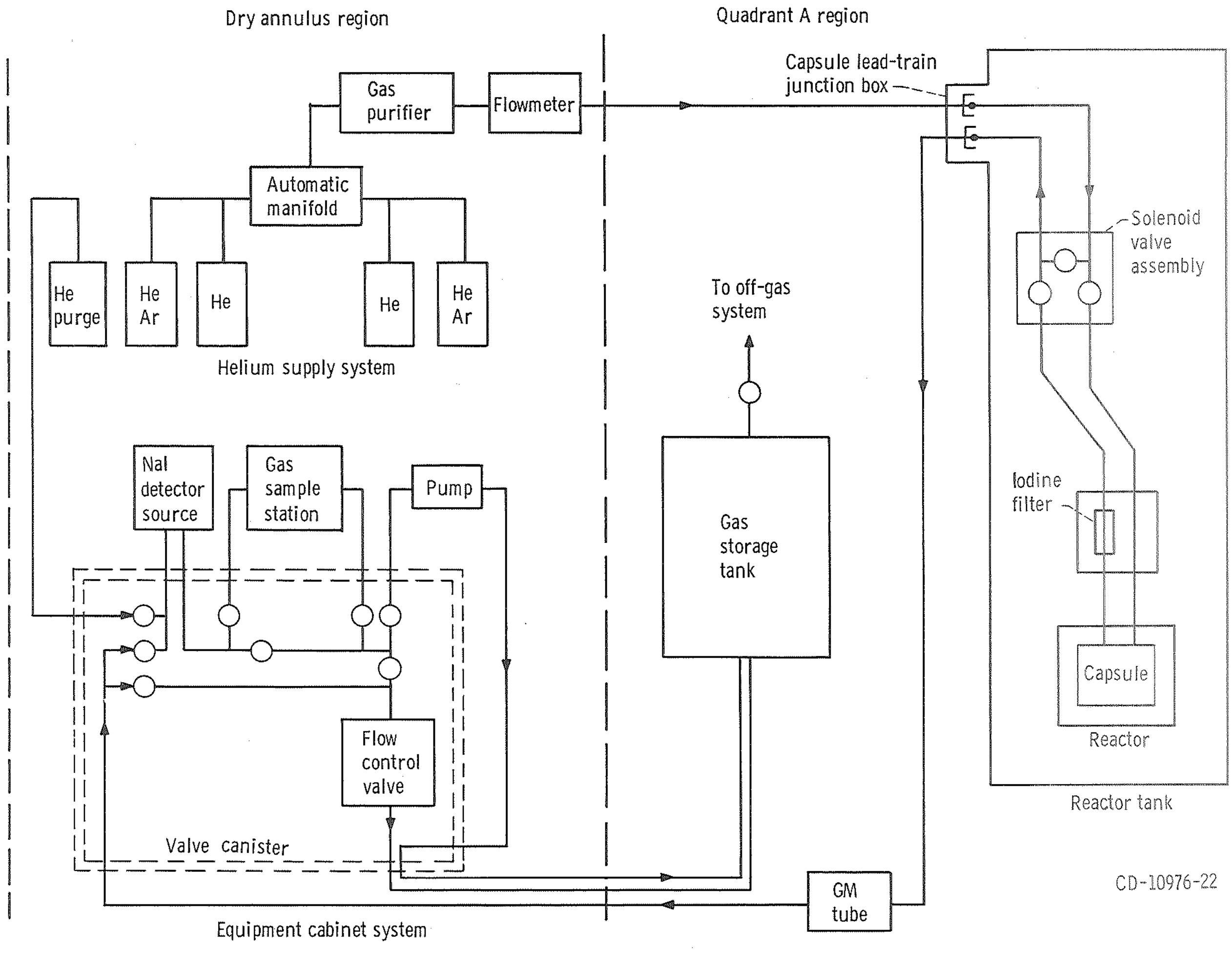


Figure 1. - Simplified sweep gas experiment flow schematic.

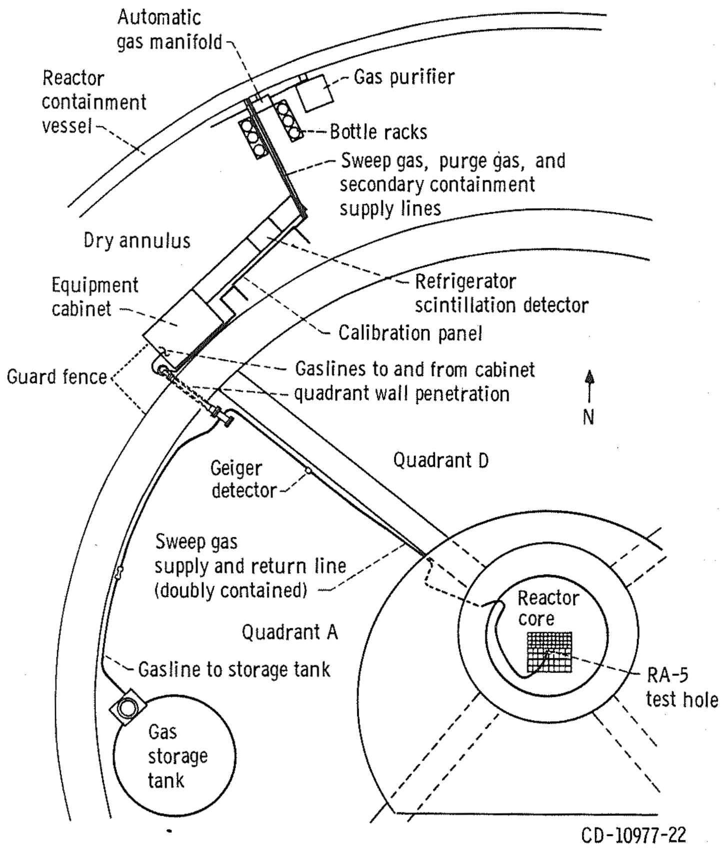


Figure 2. - General location of facility (plan view).

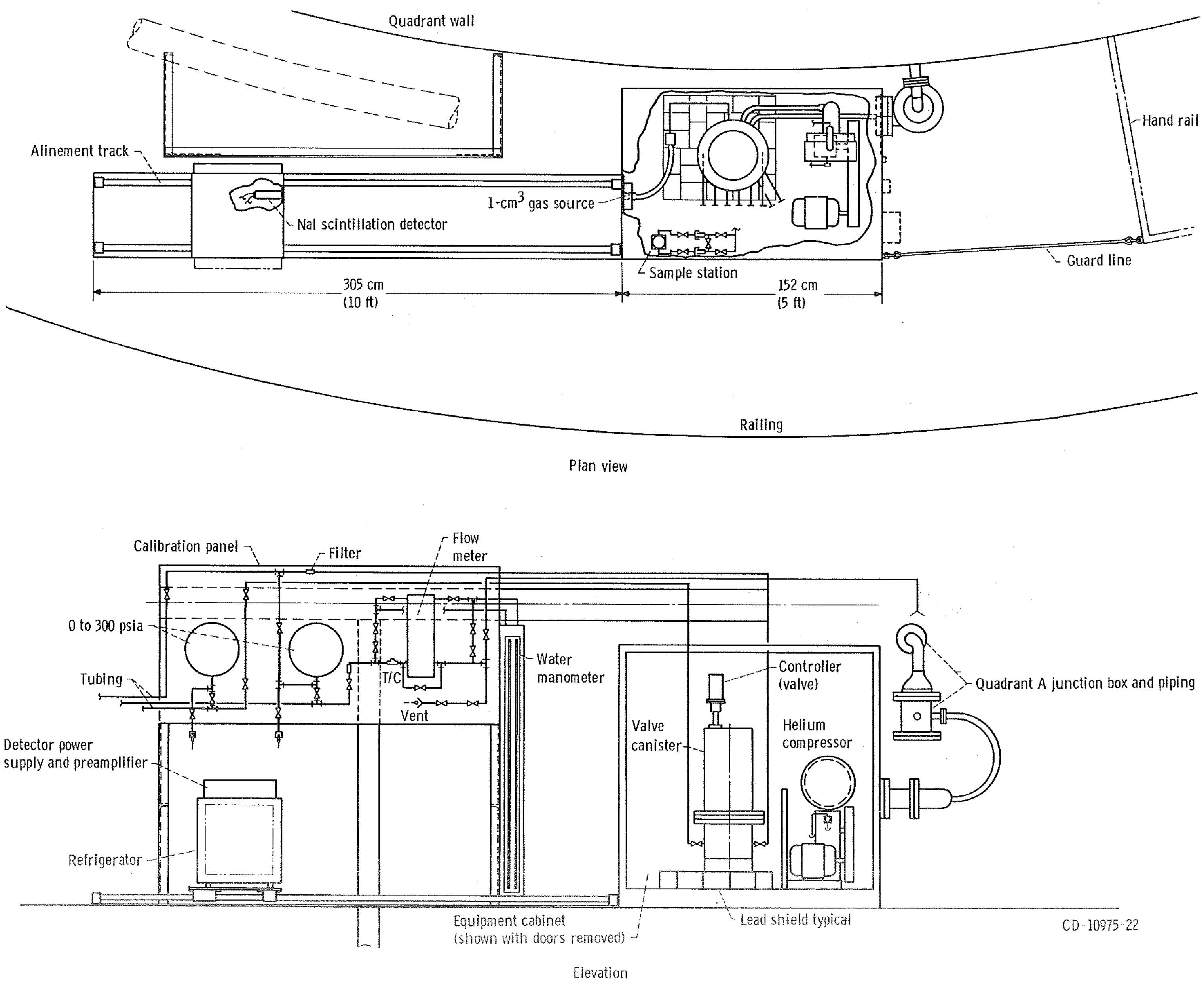


Figure 3. - Facility equipment cabinet station.

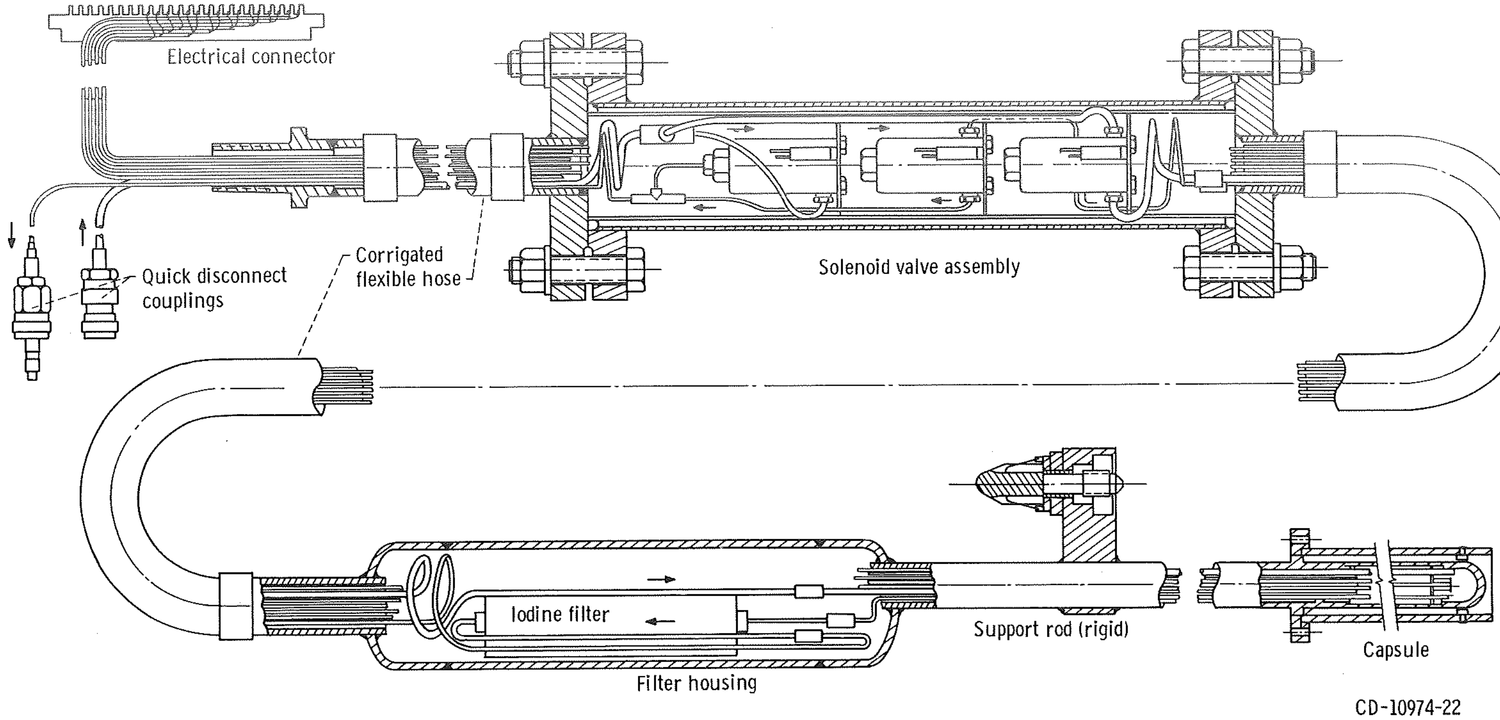


Figure 4. - Reactor tank sweep gas system.

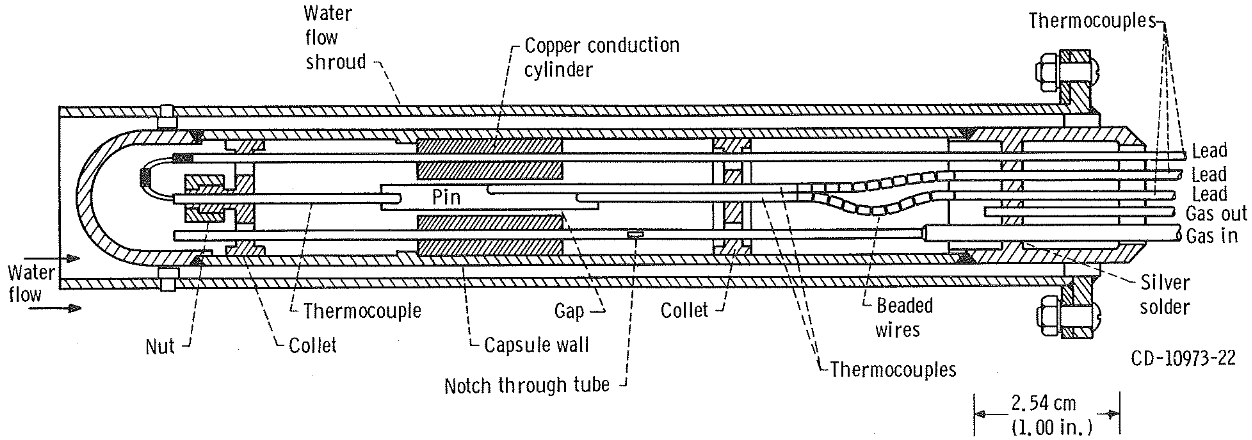
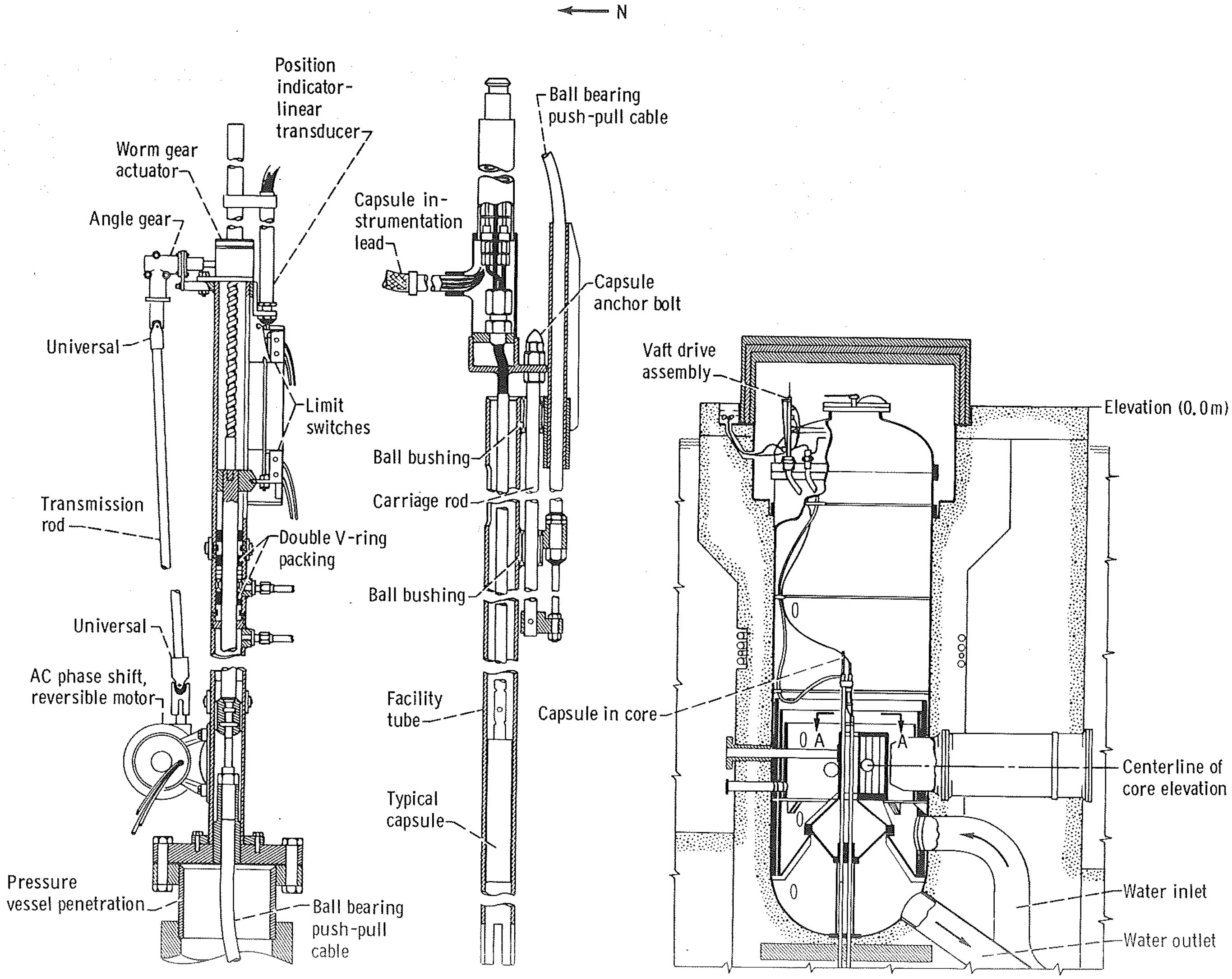


Figure 5. - Irradiation capsule.



Vaft drive assembly and pressure vessel penetration

In-core facility tube with typical capsule in place

Reactor pressure vessel with typical capsule and vaft installed

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Figure 6. - Typical VAFT installation.

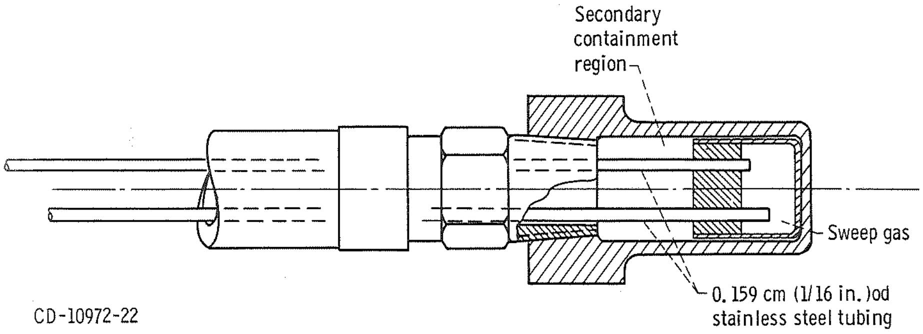

Figure 7. - Gas source for scintillation detector. Material, stainless steel; volume, 1 cubic centimeter; line of sight wall thickness, 0.127 centimeter (0.050 in.).

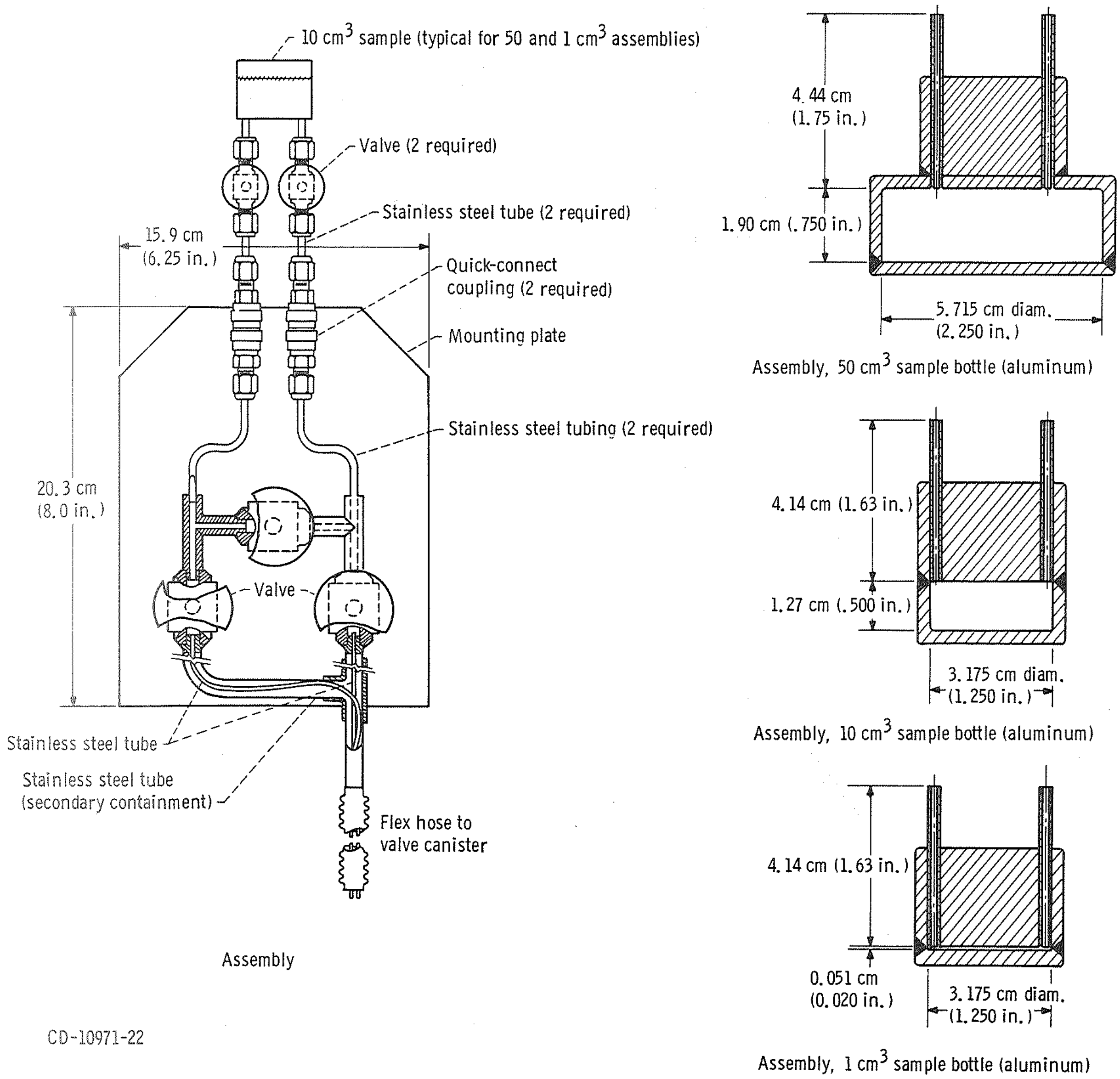

Figure 8. - Sampling station.

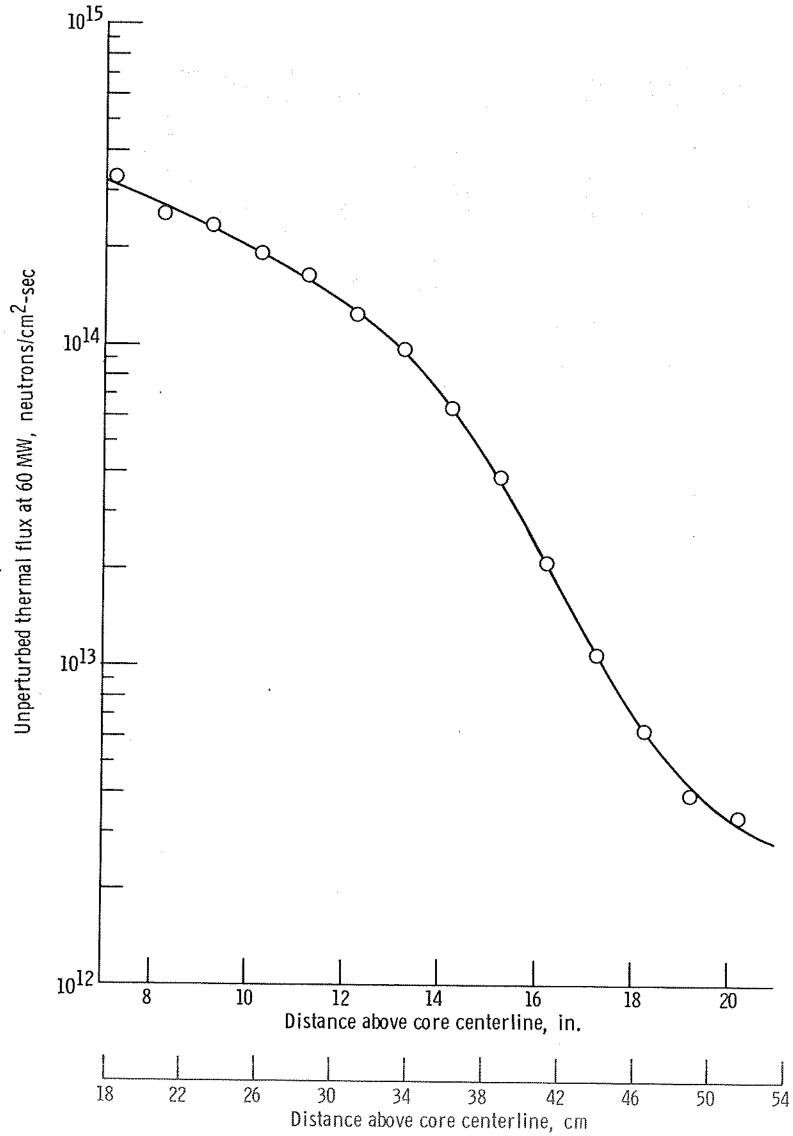

Figure 9. - Unperturbed thermal neutron flux in RA-5 reactor test hole. Values are based on mockup reactor (MUR-G) measurements with rod bank height of 40.51 centimeters (15.95 in.).

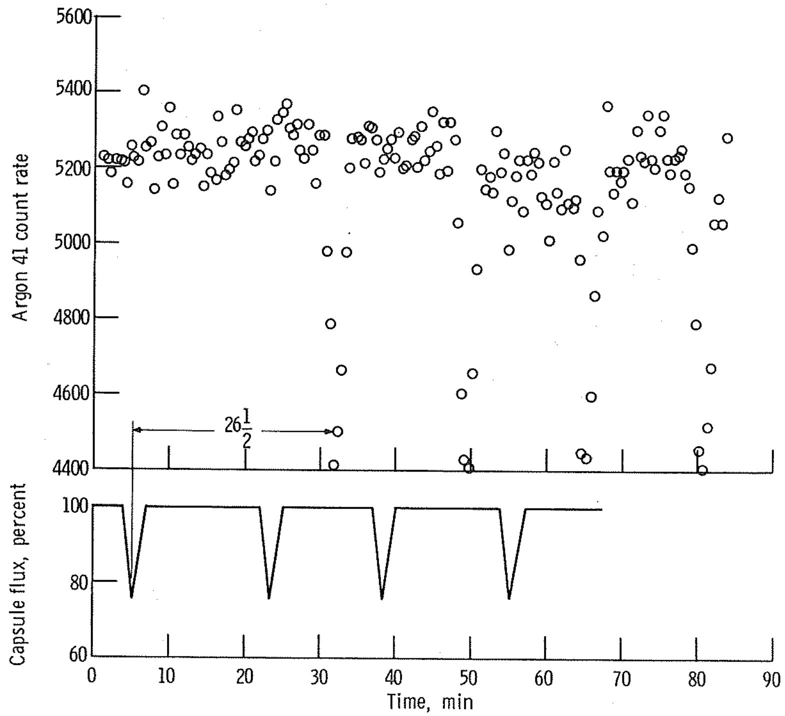

Figure 10. - Typical travel time test data. Flow rate, 0.70 cubic centimeter per second; average travel time, 26.2 minutes.

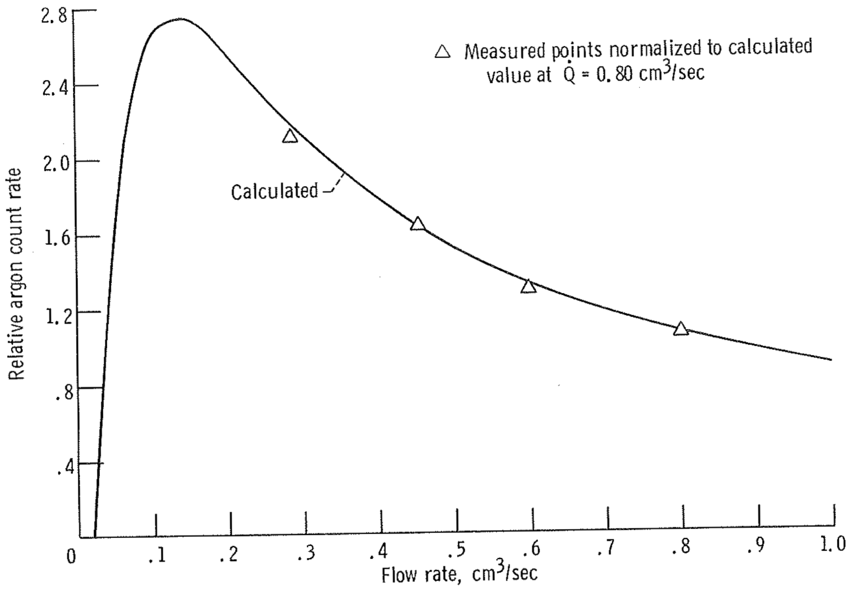

Figure 11. - Variation of argon 41 count rate with volumetric flow rate. Count rate $\propto e^{-\lambda\tau}/\dot{Q}$ where $\lambda = 1.052 \times 10^{-4}$ sec^{-1} and V = 1130 cm^3.

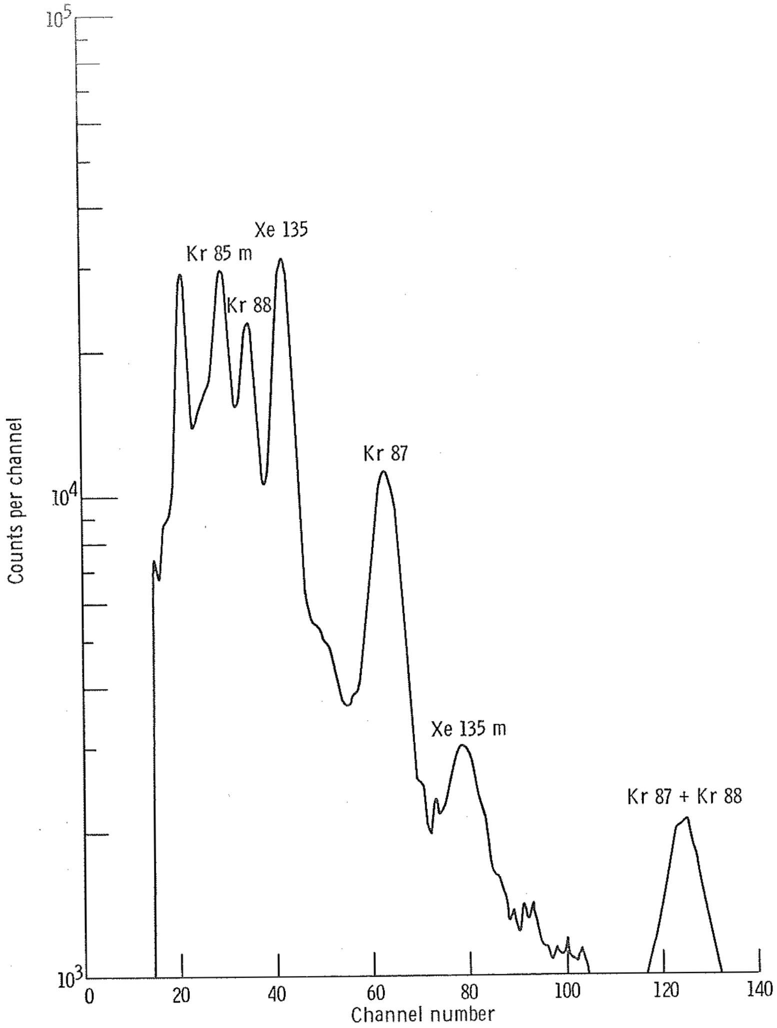

Figure 12. - Scintillation detector - fission gas gamma ray energy spectrum. Energies to 1.0 MeV.

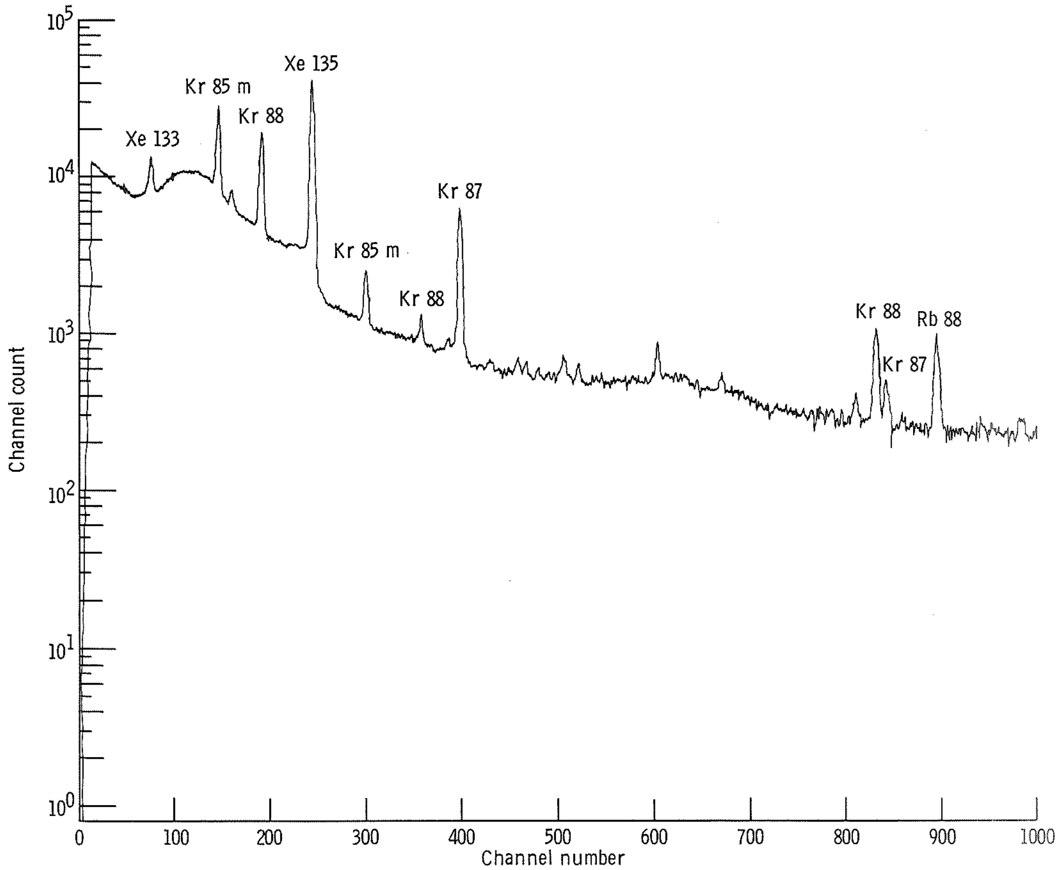

Figure 13. - Ge(Li) detector - fission gas gamma ray energy spectrum. Energies to 1.0 MeV.

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